

Process Tolerances in Roll-to-Roll Manufacturing of CIGS-Based Photovoltaics on Flexible Substrates

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ABSTRACT

Two- and three-stage co-evaporation have come to be viewed as benchmark laboratory methods for $\text{CuIn}_x\text{Ga}_{1-x}\text{Se}_2$ absorber deposition. Although quite successful and relatively easy to implement on a small R&D scale, scale-up to a commercial level proves to be challenging. Yet, large area, continuous manufacturing processes represent the most economically attractive path for thin-film PV commercialization. Large area, continuous processes necessarily differ substantially from laboratory methods, and direct process transfer is not feasible. For implementation of viable, large scale PV manufacturing methods on low-cost substrates it is necessary to understand the tolerance of the established laboratory processes to variations in deposition procedures, as they apply to low-cost roll-to-roll processing onto lightweight stainless steel foils. The success of this approach is impressively demonstrated as Global Solar Energy has achieved 90% production yield and an average production efficiency of 10%, with peak efficiencies exceeding 13% at the large-area cell level.

1. Objectives

As a Technology Partner to NREL under the Thin-Film Photovoltaics Partnerships Program the purpose of this subcontract was to move CIGS photovoltaics toward large-scale, cost-effective, manufacturing by examining the tolerance of three-stage deposition to changes in processing variables consistent with continuous RTR manufacturing at Global Solar Energy (GSE). The definition and resolution of process tolerance issues, as well as performance increase and drop in manufacturing cost satisfy many of the goals identified as the primary goal for thin-films in the Department of Energy's *Solar Energy Technologies Program: Multi-Year Technical Plan*.

2. Technical Approach

Concurrent with developing improved process control capabilities under a PVMaT subcontract, the approach under the TFPPP subcontract centered on first establishing fundamental process sensitivities of CIGS depositions onto stationary substrates. Process conditions encountered during the accelerated RTR deposition rates in GSE's production scale equipment were mapped into the laboratory equipment. Sensitivities found to have implications for RTR manufacturing were subsequently applied to the production line at Global Solar Energy. Costs and benefits of related process changes were analyzed. This work contributed toward GSE, NREL, and ITN being honored with a 2004 "R&D100 Award".

3. Results and Accomplishments

3.1 Ga/(In+Ga) profile impact on yield and efficiency

Tests were conducted to understand the tolerance of the CIGS process for composition variations and to identify hardware and process setpoint modifications to improve the output tolerance for these variations. The focus was the Ga/(Ga+In) profile through the film thickness. In three-stage depositions conducted in bell jars, virtually any Ga/(Ga+In) profile can be achieved through independent control of the Ga and In fluxes to the fixed substrate. In the production roll coaters, however, limitations in achievable profiles are imposed by the nature of the continuous roll coating process. Discrete effusion sources for each element at fixed locations are specifically responsible. Modifications were made to the standard effusion sources to provide better mixing of the elemental fluxes. The location of these modified effusion sources and their respective elemental fluxes were the test variables. Six tests consisting of 20 conditions spread over 800 ft. each were conducted. Tests were designed using standard matrices for 2 variables, 3 levels each with single or double replicates. Analysis was conducted on 36 large area cells per condition. A summary of the six test lots is shown in Table 1. The mean maximum efficiency and best uniformity between test conditions (tolerance) was obtained for configurations B and E. V_{oc} and I_{sc} had excellent model fits within each lot. Fill factor was fit poorly, indicating that it was not strongly affected by the test parameters. A comparison between I_{sc} and V_{oc} for each test lot confirmed the superiority of configurations B and repeat of C for the $I_{sc} \times V_{oc}$ product. While configuration B appears to offer the best compromise, all of the non-standard four configurations explored are capable of achieving maximum efficiencies in the 9.6 to 10.2% range.

Table 1. Cell performance summary for test lots.

Config.	Max. η (%)	V_{oc} at Max. η (mV)	I_{sc} at Max. η (mA)	FF at Max. η (%)	Ave. η (all tests)
A	9.15	531	2090	56.7	8.48
B	9.78	559	2167	55.5	9.53
C	8.73	540	2058	54.0	7.65
D	9.61	574	1997	57.7	9.09
E	9.85	556	2177	56.0	9.49
C	10.20	544	2223	58.0	9.57

3.2 NaF precursor sensitivity

Improvements in efficiency and yield due to enhanced adhesion of the absorber to the back contact were targeted via a 4 factor, 2 level experiment including center points. Evaluation of the parameter space was conducted in two independent depositions spread over two CIGS roll coaters. The experimental portion of each deposition was 800 ft. containing 18 conditions. Following device completion 60

large area cells were removed from each of the 18 conditions. Statistical analysis under removal of outliers showed the strongest impact to be the substrate temperature during NaF precursor deposition. As illustrated in Figure 1 a significant improvement in CIGS adhesion as a function of NaF precursor deposition conditions is evident. Furthermore, optimization of the NaF precursor deposition step resulted in a more robust process and enhanced device efficiency.

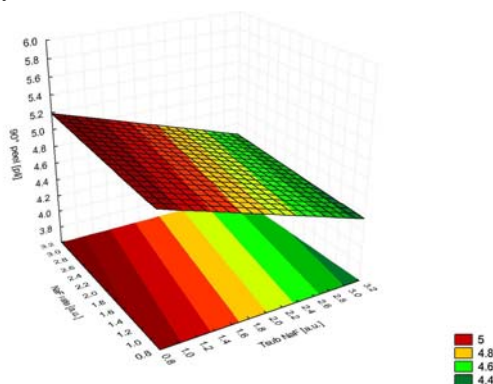


Fig. 1. Fitted surface of CIGS to back contact adhesion as a function of T_{sub} during NaF deposition and NaF rate.

3.3 Se delivery rate

In addition to the already explored process sensitivities, statistical analysis of all thin film coating processes traced inter-lot variations to the specific CIGS coaters. In section 3.1 efforts investigating the tolerance of the CIGS process to composition variation of the group III elements throughout the absorber layer have been discussed. With two of the four CIGS coaters configured to the best set resulting from this test series, screening experiments were conducted to identify the key – not directly composition driven – parameters responsible for the coater-based deviation. Experiments confirm that the process is well centered in the optimal parameter space for the variables tested. As an example, Figure 2 demonstrates the response to a change in Se delivery rate with the medium level corresponding to the current baseline process. Due to the nature of screening experiments, the width of the plateau cannot be obtained, while the design chosen still allowed for confirmation of curvature and computation of effect estimates.

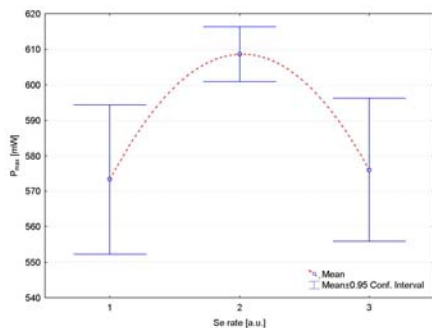


Fig. 2. Average production cell power as a function of Se rate delivery.

3.4 Feedstock purity

High-efficiency CIGS devices made in R&D laboratories are based on absorber depositions employing the highest purity source materials available. The option to utilize lower purity feedstock would benefit industry in terms of lower materials cost and better availability. However, as the impact of increasing impurity content in the source materials on device performance has not been explored, experiments to this effect were necessary to exclude potential shortfalls in device quality. Statistical analysis of an extensive device data set, as well as QE analysis, revealed the majority of the efficiency loss in the lower purity samples to be due to a shortfall in J_{sc} (mostly in the red portion of the spectrum) while V_{oc} and FF seem unaffected by source purity. Significant differences were only obtained for the low purity device set, while no statistically significant gain was evident when switching from medium quality metals to the highest purity level.

4. Conclusions

Prior to this three-year program, GSE had demonstrated average large area production device efficiencies in the 7-8% range with best devices greater than 10%. Goals under this TFPPP subcontract were to raise both numbers by 2 percentage points. Pre-contract production yield was 20% and was to be increased to 80%. All work proceeded as planned; production yield was improved to 90% while the average production efficiency has been raised to 10%, with peak efficiencies exceeding 13%.

ACKNOWLEDGEMENTS

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